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Scanning Tunneling Microscopy Studies of Heteroepitaxial GaN Thin Films Grown by Plasma-assisted Molecular Beam Epitaxy

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III-V nitrides, including GaN, AlN, InN and their alloys, with wide bandgap and stable physical and chemical properties, have been successfully applied to fabricate blue light emitting diodes (LEDs) and laser diodes (LDs) recently. Despite the significant technological success, our understanding of their fundamental properties, such as growth mechanism and surface structure, is still rather limited. In the present study, we, by using scanning tunneling microscopy (STM), reflection high energy electron diffraction (RHEED), and first-principles total-energy calculations, have carried out a systematic investigation of surface reconstructions occurred on the GaN thin films, which are grown on both hexagonal 6H-SiC and cubic GaAs substrates by plasma-assisted molecular beam epitaxy (MBE) [1--4].

SiC has advantages to be a substrate for the epitaxial growth of GaN than sapphire, due to its smaller lattice mismatch(3.5%) and closer thermal expansion coefficient. In the present work, a two-step method which combines the atmospheric hydrogen etching and UHV Si etching has been developed to prepare clean surfaces of the Si-terminated 6H-SiC(0001) substrate. Our AFM and STM measurements show that the resulting surface is scratch-free, atomically flat, containing large-area terraces separated by straight steps. 3x3 or $\sqrt{3}$x$\sqrt{3}$ reconstruction is obtained depending on the Si coverage. The surface quality is comparable with that of the well-developed Si or GaAs. As to the C-terminated 6H-SiC, similar quality of surface is obtained by UHV Si etching, and the surface exhibits 2x2 reconstruction coexisting with additional carbon cluster structure.

On the Si-terminated 6H-SiC(0001), wurtzite GaN films exhibit Ga polarity, i.e. GaN(0001), which is confirmed by interface bonding, surface reconstructions and the chemical etching (they are resistant to the molten KOH). The as-quenched GaN(0001) surface after growth usually exhibits 1x1 RHEED pattern. However, post-growth Ga deposition followed by annealing at low temperature(200~400°C) results in the formation of a number of reconstructions with smooth morphology, which has allowed us study them by STM. These reconstructions are 2x2, 4x4, 5x5, 5$\sqrt{3}$x2$\sqrt{13}$, $\sqrt{7}$x$\sqrt{7}$, 10x10, and '1x1' Ga-fluid. Among them, the 2x2 and 4x4 have the similar features, and their atomic structure is interpreted in term of a T_4 Ga adatom model based on the atomically-resolved STM images and first-principle total energy calculations. All other Ga-stabilized structures can be understood under this Ga adatom scheme. We note that some phases, such as 5x5, 5√3x2√13, made of one-dimensional linear chain structure, can be explained by the Peierls distortion due to many-body effects. The auto-compensation, one of the guiding rules for conventional III-V compound semiconductor surface reconstruction, may not be strictly satisfied on the GaN surface.

On the C-terminated 6H-SiC$(000\overline{1})$, our results for the first time experimentally demonstrate that the GaN film has N polarity, i.e. GaN$(000\overline{1})$, displaying a different set of Ga-stabilized reconstructions compared with that of GaN(0001) surface. Therefore, wurtzite GaN films grown on the Si face of 6H-SiC have Ga polarity, while on the C-face of 6H-SiC the N polarity. The key point for this polarity correlation between the polar GaN and polar SiC substrate is the preferential bonding of N-Si, Ga-C/Al-C at the interface. On the GaN$(000\overline{1})$ surfaces, with the post-growth Ga deposition, several new reconstructions, such as 2x4 and 2√7x2√7, have been identified. The atomic-scale STM images show that the 2√7x2√7 reconstruction is made of the complicated ring-like structure and displays strong bias-dependence, and tunneling spectroscopy (STS) measurement indicates it is semiconducting with small gap. We will document that 2√7x2√7 structure does not agree with the electron counting, and can be better understood by a two-dimensional charge density wave (CDW) state. In order to understand all the reconstructions on GaN surface, we have to consider many-body effects in addition to the electron counting which is based on single-particle theory.

No ordered N-rich structure has been obtained, irrespective of the film polarity. The strong cohesive energy of N_2 (5.0ev) makes the N adatoms thermodynamically unstable against evaporation as N_2 molecules, and the mobility of N atoms on both polar surfaces is poor, such that the N adatom structure predicted by theoretical studies is difficult to access kinetically.

We have also performed an in-situ STM study of the epitaxial growth of GaN thin films on the cubic GaAs(001). We found that at the initial stage, the exposure of the GaAs(001) to the reactive N species from N plasma induces a flat nitride layer with the 3x3 reconstruction and anisotropic vacancy islands. With prolonged exposure time or higher substrate temperature, the surface morphology is degraded and the 3-D islands appear. This nitridation process is found to influence significantly the subsequent GaN growth. When the growth is initiated on the flat nitride layer, the single-phase cubic crystalline GaN forms, on which the 2x2 reconstruction is identified. However, the growth from the rough nitride layer always exhibits 3-D mode, and the resulting GaN film usually contains mixed cubic and hexagonal phases. The coexistence of these two phases can be understood by the equivalence between the {111} plane of the cubic GaN and the {0001} plane of the wurtzite GaN. Possible mechanism responsible for these results is discussed.

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3. Xue, et al., **Appl. Phys. Lett. 74**, 2468 (1999).
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